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Co-conversion of minor actinides in uranium based oxidic precursors by internal gelation

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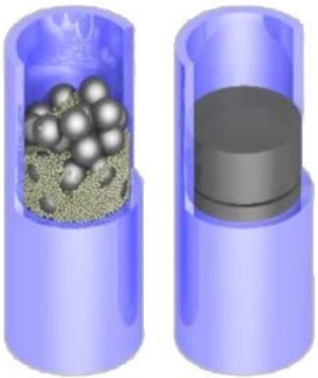
Introduction

Advanced nuclear fuels → particle fuels

- good swelling behaviour
- can directly be inserted into the fuel pin



http://asgardproject.eu/download/var/files/poster1_full.jpg

Sol-gel technique:

- transformation of a liquid sol into a solid gel

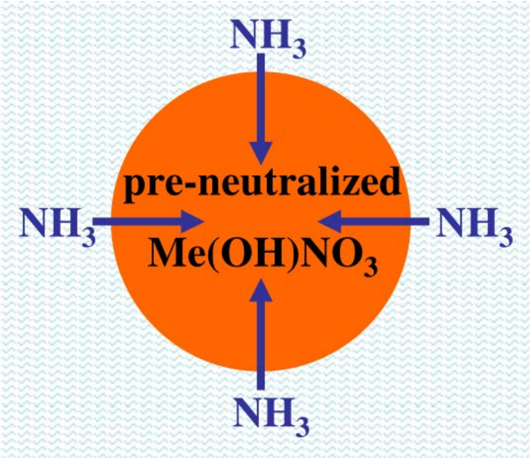
  → creation of intermolecular networks

Task:

- preparation of defined particles with defined microstructure
- U/Nd ratio adjustment (Nd^{III} as surrogate for An^{III})

Sol-gel processes

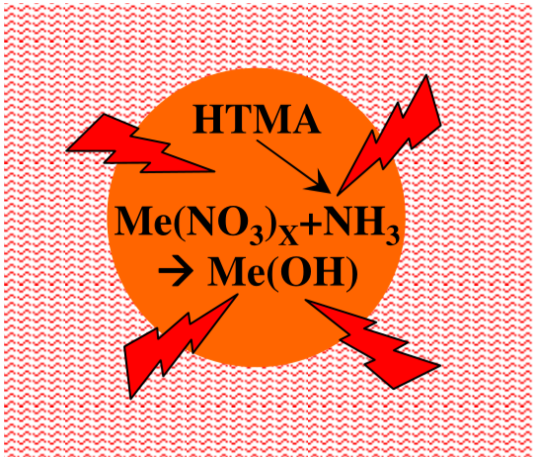
external gelation



mass transfer of ammonia

- no heat carrier required

internal gelation

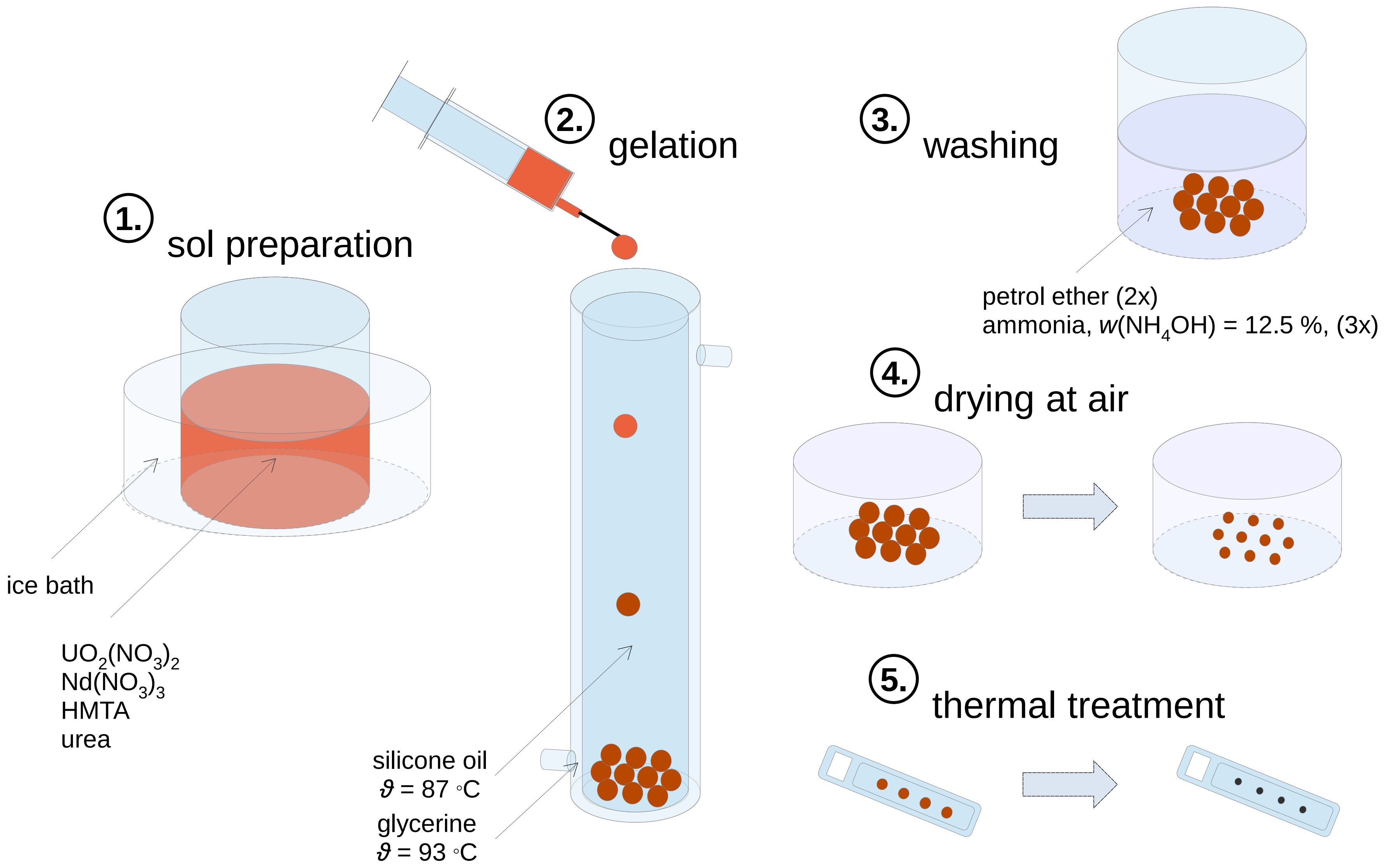


heat transfer

- co-processing of different metals
- wide range of sphere sizes
- capable for non oxide ceramics (nitride, carbide)
- complex sol-formulation

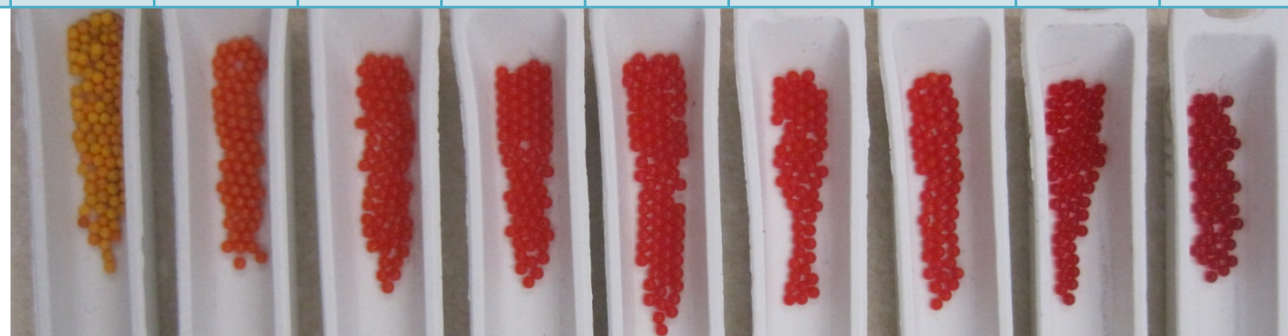
M. Pouchon et al. *Sphere-Pac and VIPAC Fuel*, In R. J. Konings et al. (eds.) *Comprehensive Nuclear Materials, V*ol. 3 (11), pp 275–312., 2012.

Internal-gelation procedure



Particle preparation

$\chi(\mathrm{Nd})_{\mathrm{ICP\text{-}MS}}$	%	0	5.80	11.99	17.40	22.62	27.59	33.49	37.68	42.63
$c(\mathrm{U+Nd})_{\mathrm{sol}}$	mol/L	2.50	2.60	2.62	2.56	2.58	2.60	2.62	2.65	2.67
$\bar{m}(\mathrm{particle})$	mg		5.38	5.23	4.96	5.24	4.83	5.00	5.34	5.55
$\bar{d}(\mathrm{particle})$	mm		1.35	1.35	1.33	1.38	1.34	1.35	1.38	1.41
$\bar{\rho}(\mathrm{particle})$	g/cm^3		4.15	4.04	3.99	3.85	3.87	3.86	3.85	3.81

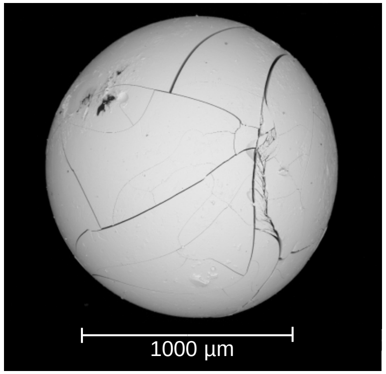


$$\chi(\mathrm{Nd}) = \frac{n(\mathrm{Nd})}{n(\mathrm{Nd+U})} \qquad R(\mathrm{urea}) = \frac{n(\mathrm{urea})}{n(M^{n+})} = 1.80 \qquad R(\mathrm{HMTA}) = \frac{n(\mathrm{HMTA})}{n(M^{n+})} = 1.35$$

Schreinemachers et al., *Progress in Nuclear Energy, Elsevier,* 2013
“Characterization of uranium neodymium oxide microspheres synthesized by internal gelation“

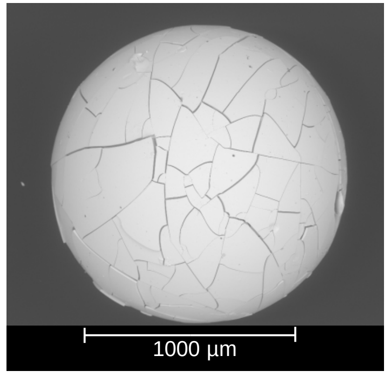
Particle characterization by SEM

$\chi(Nd)$ = 0 %



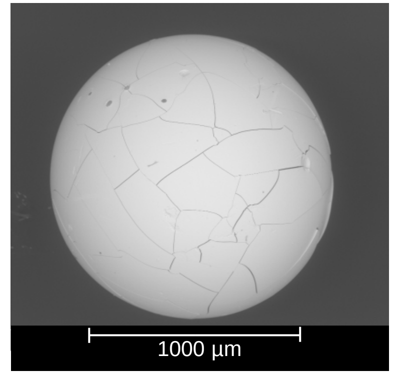
$\bar{d}$ = 1.356 mm

$\chi(Nd)$ = 5.80 %



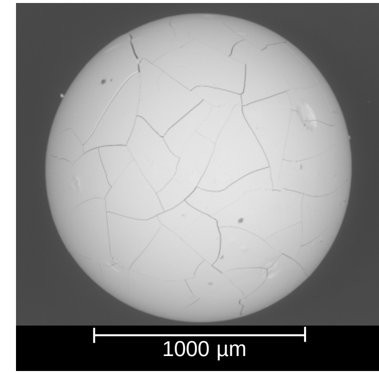
$\bar{d}$ = 1.323 mm

$\chi(Nd)$ = 27.59 %



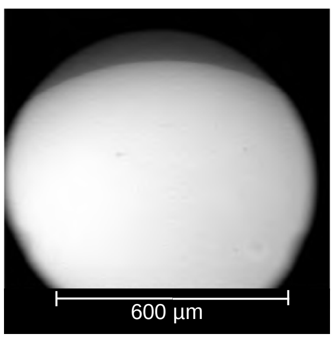
$\bar{d}$ = 1.263 mm

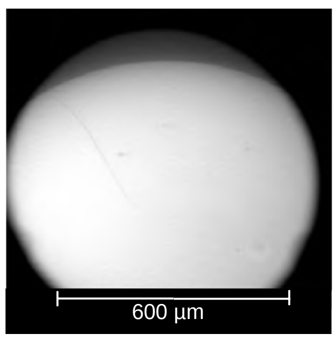
$\chi(Nd)$ = 42.63 %

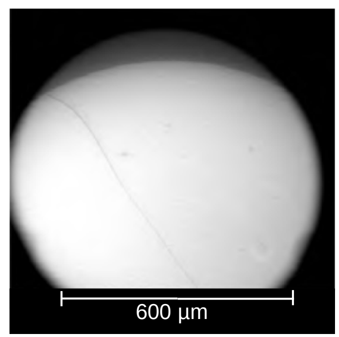


$\bar{d}$ = 1.385 mm

- **ESEM analysis of the green bodies**







($\chi(Nd)$ = 22.6 %, 50°C, 850 Pa, humidity: 31.6 %)

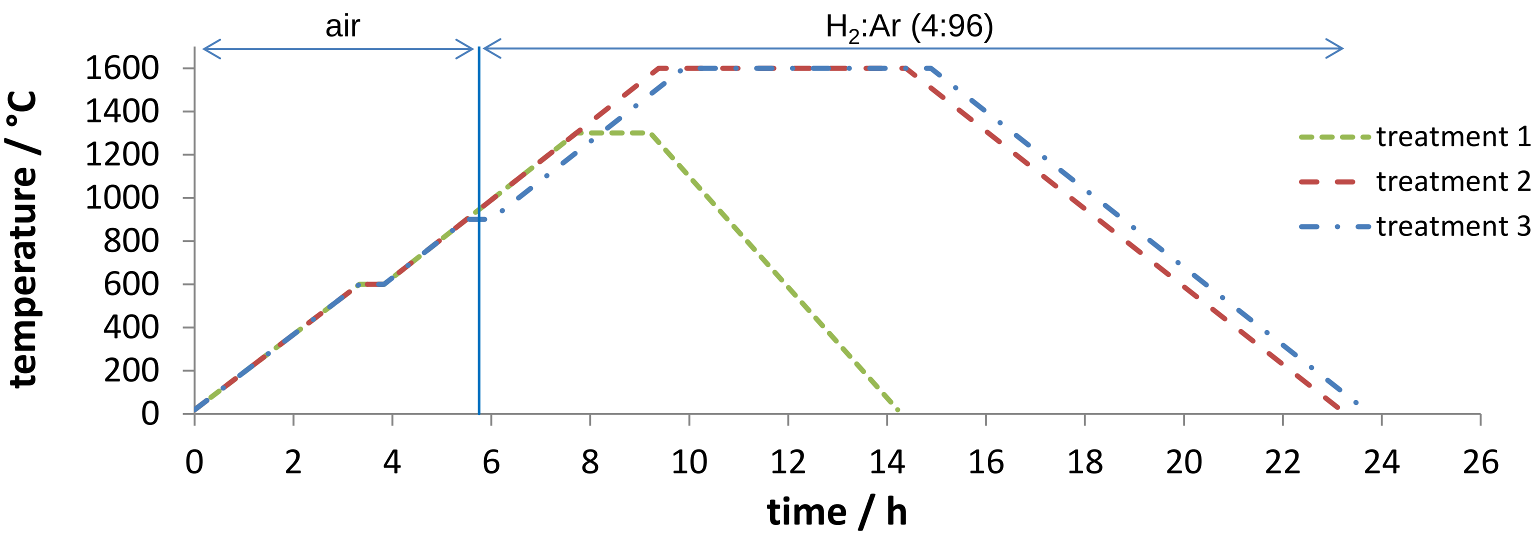
initial conditions
5 °C, 97.0 % humidity, 850 Pa

temperature increase
5 °C → 50 °C, 5 $\frac{°C}{min}$

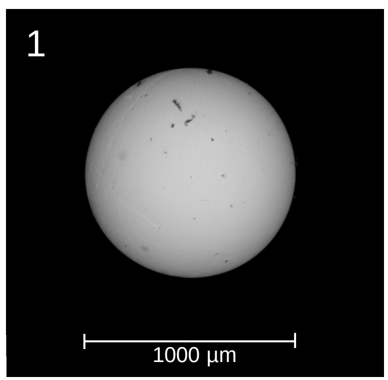
pressure decrease
850 Pa → 70 Pa, 3.0 % humidity

Schreinemachers et al., *Progress in Nuclear Energy, Elsevier,* 2013
“Characterization of uranium neodymium oxide microspheres synthesized by internal gelation“

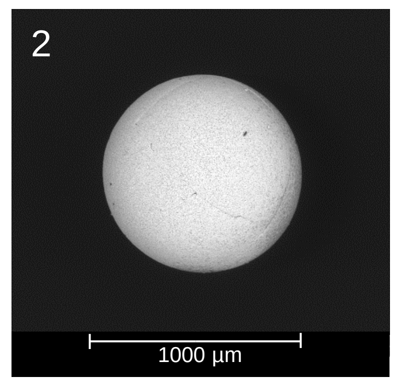
Thermal treatment



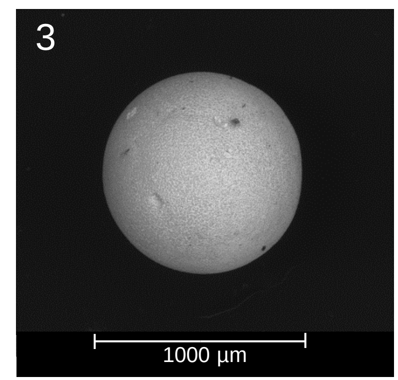
χ(Nd) = 42.63 %



$\overline{m}$ = 4.17 mg
$\overline{d}$ = 919.1 µm
$\overline{\rho}$ = 10.25 g/cm³



$\overline{m}$ = 3.96 mg
$\overline{d}$ = 899.7 µm
$\overline{\rho}$ = 10.13 g/cm³



$\overline{m}$ = 3.96 mg
$\overline{d}$ = 922.3 µm
$\overline{\rho}$ = 9.56 g/cm³

Schreinemachers et al., *Progress in Nuclear Energy, Elsevier,* 2013
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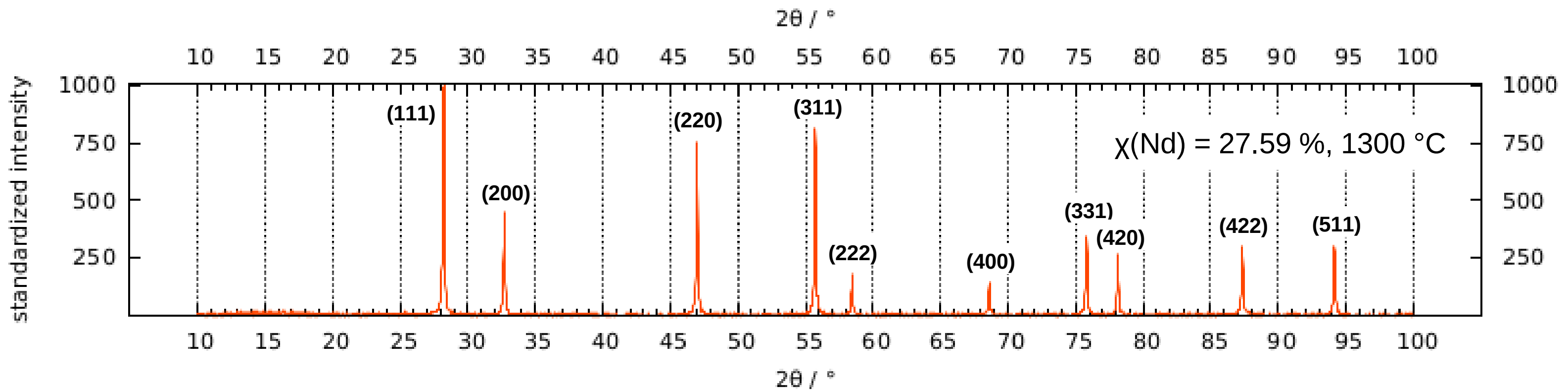
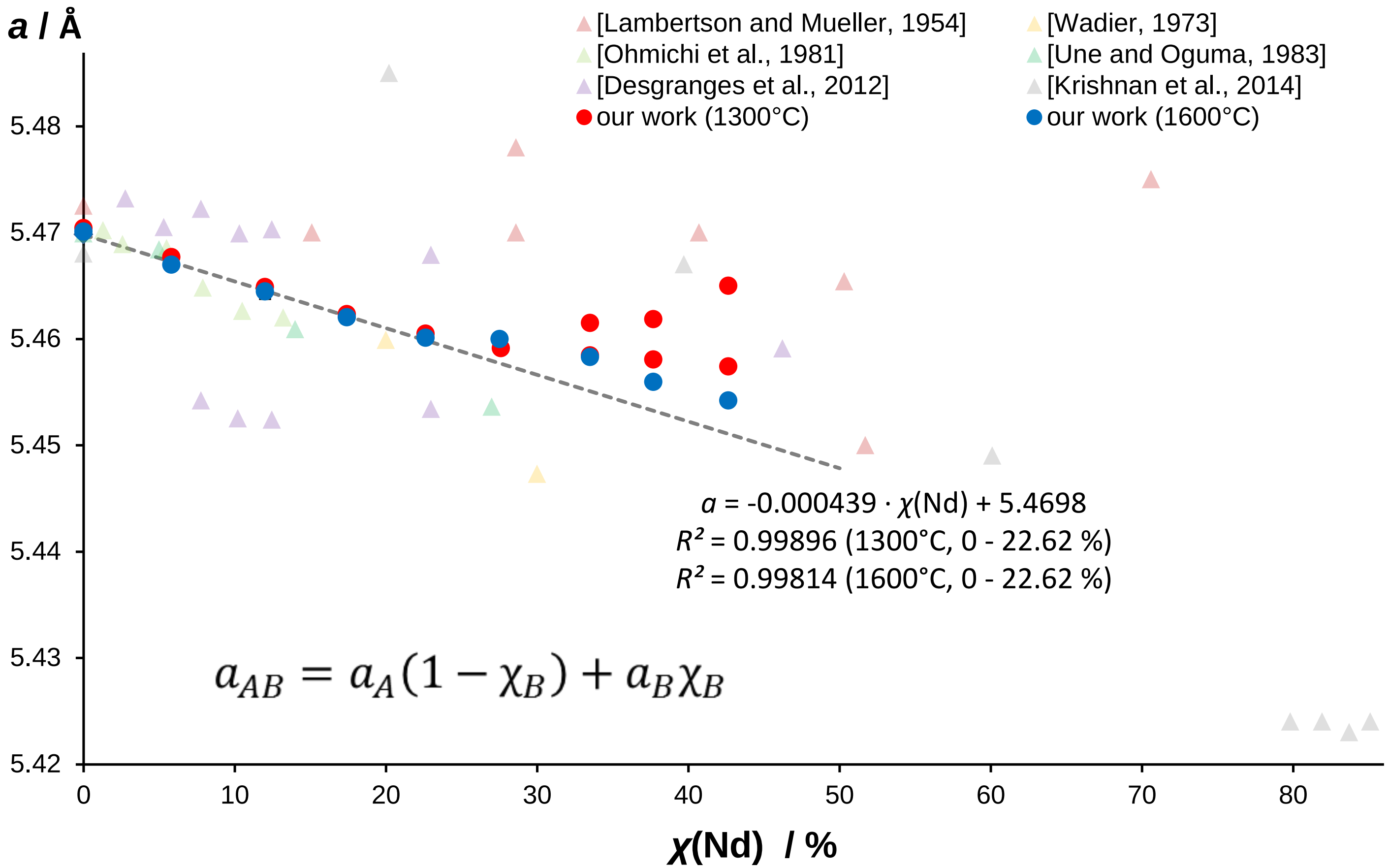
XRD analysis



- crystalline particles
    - fluorite structure
    - $Nd_xU_{1-x}O_{2\pm y}$
- higher *2θ* values with increasing Nd content
- 1300°C → two phases
- 1600°C → one phase
- lattice parameter *a*
- mean crystalline size *L*
- lattice distortion |*ε*|

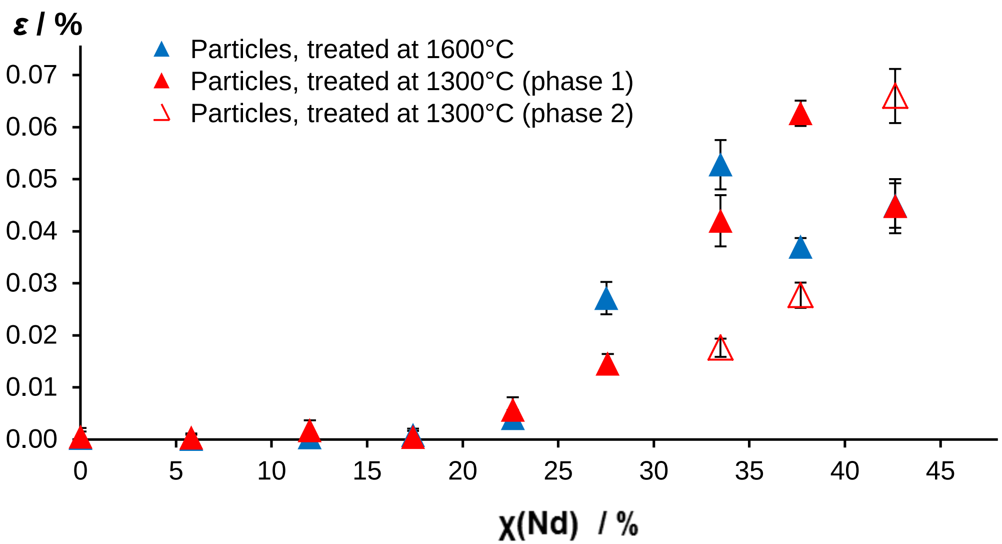
Lattice parameter

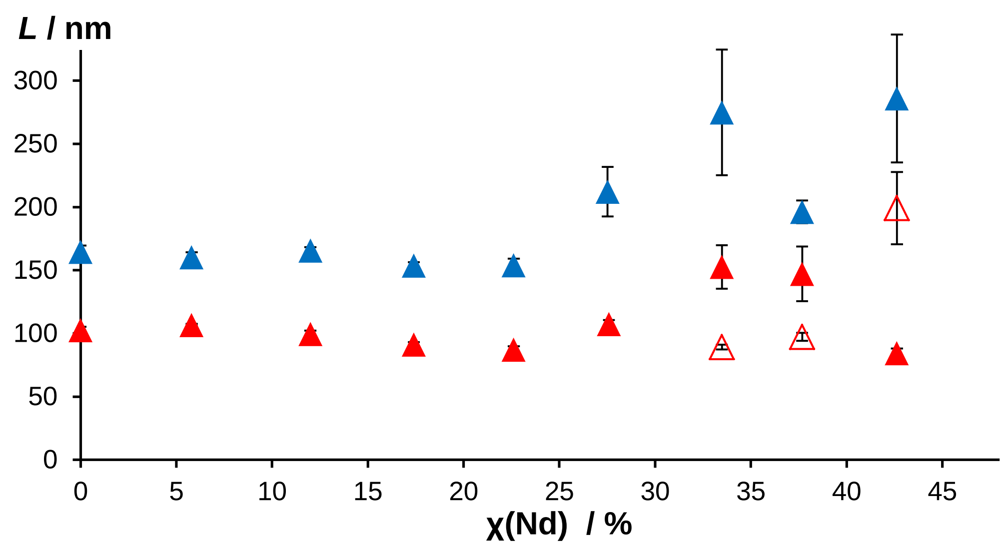


$$a_{AB} = a_A(1-\chi_B) + a_B\chi_B$$

→ lattice formation depends on synthesis route

Lattice distortion / mean crystalline size





- *ε* almost 0 up to 25 % Nd
- correlation of deviation in lattice parameter and *ε*
- *L* depends on sintering time and temperature
- 1600°C → ~ 160 nm
- 1300°C → ~ 100 nm

Conclusion

- *R*(urea) = 1.80 & *R*(HMTA) = 1.35 sufficient for the preparation of UO_2 particles with Nd contents from 0 % to 42.63 %.
- Average particle mass and diameter with small standard deviation, although the process was manual.
- Spherical shape proven by SEM and optical microscopy.
- One cubic phase for particles sintered at 1300°C in H_2:Ar up to χ(Nd) = 27.59 %, higher contents lead to two phases.
- Single phase UO_2/Nd_2O_3 ceramics for the whole observed Nd range sintered at 1600°C in air and H_2:Ar.
- Equilibrium solid solutions of the sensitive UO_2/Nd_2O_3 system can be fabricated by the internal gelation synthesis route.

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Thank you for your attention!